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AEC RESEARCH AND
DEVELOPMENT REPORT

GA-5487

ADVANCED BERYLLIUM OXIDE CONCEPTS
PROGRESS REPORT
FOR THE QUARTER ENDING JUNE 30, 1964

Prepared under
Contract AT(04-3)-187
for the
San Francisco Operations Office
U.S. Atomic Energy Commission

August 1, 1964

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GENERAL ATOMIC
DIVISION OF
GENERAL DYNAMICS

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PREVIOUS REPORTS IN THIS SERIES

GA-3783-October, November, December, 1962

GA-4165-January, February, March, 1963

GA-4451-April, May, June, 1963

GA-4710-July, August, September, 1963

GA-4966-October, November, December, 1963

GA-5036-January, February, March, 1964

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1. INTRODUCTION AND SUMMARY

The Experimental Beryllium Oxide Reactor (EBOR) Test Module Program is supported by the U.S. Atomic Energy Commission under Contract AT(04-3)-187 and has the following objectives:

1. To identify reactor concepts that effectively utilize the nuclear characteristics of beryllium oxide.
2. To determine the nuclear, heat-transfer, and major mechanical-design features of reactor modules applicable to beryllium oxide advanced reactor concepts.
3. To investigate the behavior of materials that have potential applications in advanced concepts.
4. To specify experimental elements for EBOR that incorporate a maximum number of significant design features of advanced concepts.

Parametric work continued in the two specific areas of interest: investigations of thorium utilization and plutonium utilization. In addition, work proceeded on the preliminary design of the first EBOR test module.

THORIUM UTILIZATION INVESTIGATION

Work on possible alternative fuel-element designs continued during the quarter. In particular, interest was centered on the potential use of segmented annular fuel bodies in place of the single annular fuel ring. For small ratios of thermal conductivities of fuel and fertile material to those of matrix material, it was found that the more finely the fuel body is subdivided into small sections within the solid fuel element, the lower the maximum fuel temperature becomes for a fixed power output from the element. Some calculations were also performed on an alternative design consisting of a cylindrical element cooled both internally and externally and having a fueled inner spine. A higher power density appears possible with this type of element, and since the fuel region is essentially segmented into several regions in any single element, fewer elements would be required for a given power output. Complications in manufacture could possibly be the major disadvantage of this type of element.

In the materials area, the four-region sweep capsule was reinserted in the Materials Testing Reactor (MTR) on May 18 after the leak in the lead tube was repaired. Two weeks after reinsertion, however, another leak developed in the vicinity of the samples. The capsule was permanently discharged from the reactor at the end of the quarter. Discussions have been initiated concerning the ultimate disposal of this capsule. No decision has as yet been made on the question of whether sufficient irradiation at temperature has been accumulated to merit a postirradiation examination of the samples.

During the quarter, the analytical procedure for the determination of Li^6 in beryllia was improved. Using the improved procedure it was found that the Li^6 concentration in the irradiated MGCR-2 samples agreed favorably with the calculated values. Further work is underway to determine the diffusion coefficient of Li^6 in BeO .

PLUTONIUM UTILIZATION INVESTIGATION

The preliminary physics investigation of the feasibility of a plutonium-fueled second core for EBOR was completed. As reported in the previous quarterly progress report, the feasibility of a core with power limited to 10 Mw appeared to depend entirely on the use of little or no metal (as cladding) in the core. This result was confirmed by the calculations completed during the quarter, and some results of the more detailed examination are presented herein.

Work continued on the investigation of silicon carbide as a possible cladding material. Investigations were initiated on two types of silicon carbide material: extruded SiC as manufactured by Carborundum Company and vapor-deposited SiC as made by San Fernando Laboratories.

EBOR TEST MODULE

Work in this area was concentrated on establishing the basic design parameters for the first element for testing in the EBOR Test Module Program. This test module will be used to investigate the feasibility of a UO_2 - ThO_2 fuel cycle utilizing BeO as moderator and fuel diluent. As noted previously, graphite has been established as the cladding for the reference element. The possible damage caused by the oxidation of the graphite during emergency air cooling of EBOR was investigated. The analysis indicated that the heat flow from the burning element did not result in dangerously high temperatures or temperature gradients in the adjacent EBOR elements. Other possible perturbations to the EBOR core are still under investigation. In regard to the test element itself, a clearer picture of the effect of the gap between the fuel body and the graphite sleeve was obtained.

2. THORIUM UTILIZATION

ENGINEERING

Alternative Fuel-element Design

This fuel element, which was previously considered in GA-5036, is represented on Fig. 1. It consists of a cylindrical matrix in which a certain number of holes are either drilled or extruded. These holes are occupied by fueled material, such as UO_2 and ThO_2 mixed with BeO . The number of such holes may vary, but they are supposed to occupy a fixed volume fraction. Their shape may vary from flat segments, forming an annular ring at the limit, to elongated pie-shaped holes extending almost to the center. In all cases, the outer border of the fuel compact remains at the same distance from the cooled outer surface of the fuel element. It was reported in GA-5036 that the maximum internal temperature of an externally cooled cylindrical fuel element generating a given amount of heat could be decreased substantially by fractioning the annular fuel compact. This result is valid for small ratios of thermal conductivities of fuel (and fertile material) to matrix material (for example, heavy-metal oxides and BeO) and for a fixed fractional volume of fuel element occupied by the fuel (20% in the present studies). The sleeve material was assumed to have the same thermal conductivity as the matrix. Large numbers of fuel-containing holes produced better results.

Figure 1 shows the variation of the relative difference in maximum internal fuel-element temperature with respect to an annular ring. This relative decrease in maximum temperature is plotted versus the number of holes containing fuel and versus the square of the ratio of the inner radius of the hole R_0 to the inner radius of the annular compact R_a . T_a represents the maximum temperature when the fuel is concentrated in an annulus. The extreme left side of the diagram, which corresponds to a small departure from an annular fuel compact, has been studied in detail both analytically and by machine computations. It is seen that a 10% angular opening in the annulus ($\theta/\theta_m = 0.9$) corresponds to a relative decrease in maximum temperature, which varies from 10% for 6 holes to about 20% for 36 holes. Since the difference between maximum internal fuel-element temperature and bulk coolant temperature (T_a) may be as large as 1000°C , a 10% opening could bring a decrease in maximum temperature of 100° to 200°C . Figure 2 shows a plot of the isotherms for $n = 8$ (where n is the number of holes) and $\theta/\theta_m = 0.9$; the bulk coolant temperature is taken to be zero

Fig. 1--Relative maximum fuel-element temperature versus geometry and number of holes for a ratio of matrix and fuel conductivities of 10

Fig. 2--Isotherms for 8 holes, $\theta/\theta_m = 0.90$, and a ratio of thermal conductivities of 10

and the surface Biot* number, $h R_2 / K_2$, is equal to 2. The ratio of conductivities is 10, and 20% of the fuel element is occupied by the fuel. As compared with a complete annulus, the maximum temperature moves inward inside the fuel compact, since the inner face of the fueled region is not insulated any more. Some heat is transferred through the spine and the opening in the annulus toward the surface cooled by the gas. For a given relative opening $[1 - (\theta/\theta_m)]$, more of the compact is affected when its length-to-thickness ratio is small. This effect is seen on Fig. 1, where the decrease in maximum temperature is larger for larger numbers of holes.

Figure 1 shows that the maximum temperature keeps decreasing with increasing opening $[1 - (\theta/\theta_m)]$ up to approximately $1/2$ for large numbers of holes (24 to 36), while it starts increasing again for values of $[1 - (\theta/\theta_m)] \geq 0.10$ to 0.20 for smaller numbers of holes (6 to 12). An explanation of this phenomenon may be seen on Figs. 3 and 4 for $n = 8$. For $\theta/\theta_m = 1/2$, as shown on Fig. 3, the fueled region is relatively thick in both directions and the maximum temperature is about the same as for the annulus. In Fig. 4, where $\theta/\theta_m = 0.69$, the fuel compact is very elongated and most of the heat flows circumferentially out from the hot spot through a relatively thin region; the maximum temperature is much lower than for the annulus and is about the same as for $\theta/\theta_m = 0.9$.

The same behavior is observed for $n = 6$, but the square holes are thicker and therefore much worse than for annular compacts; in that case, for $n = 6$, $\theta/\theta_m \cong 0.90$ is nearly optimum. For $n = 12$, the annulus yields the highest maximum temperature (see Fig. 1) and the maximum temperature does not vary much between $\theta/\theta_m = 0.90$ and 0.35 , since the compacts are never very thick. For larger numbers of holes, 24 to 36, the lowest value of the maximum temperature is reached for $\theta/\theta_m \cong 1/2$. For very elongated holes $[1 - (\theta/\theta_m) \cong 0.65]$ the location of the maximum temperature is too far from the cooled surface.

Figure 5 compares the temperature distribution in circular fuel compacts and in equivalent circular holes defined as having the same length $(R_1 - R_0)$ as the circle diameter and the same surface area as the compacts. The maximum distance between fuel holes and cooled surface is the same in both cases (see Fig. 1). The maximum temperatures are shown to be only slightly higher (4%) for the true circular hole at the same heat flux. Equivalent circular holes, represented on Fig. 1, are seen to be much worse than the optimum shape for small numbers of holes, since circular holes are much too thick. For larger numbers of holes, i. e., smaller area per hole, circular holes are more comparable with optimum-shaped holes, although even for $n = 12$, the maximum relative decrease in temperature is about 18.5%, while it is only about 10% for a circular hole.

* Ratio of heat transfer by convection in fluid to heat transfer by conduction in solid.

Fig. 3--Isotherms for 8 holes, $\theta/\theta_m = 0.50$, and a ratio of thermal conductivities of 10

Fig. 4--Isotherms for 8 holes, $\theta/\theta_m = 0.69$, and a ratio of thermal conductivities of 10

NOTE:

TEMPERATURES IN ARBITRARY UNITS
ZERO COOLANT TEMPERATURE

Fig. 5--Comparison of temperature distributions in circular fuel compacts and equivalent fuel compacts ($n = 12$ and $K_2/K_1 = 10$)

Optimum Concentration in Fueled Region of Fuel Element Producing Maximum Heat Flux⁽¹⁾

In the previous quarterly progress report (GA-5036), the optimum thickness of an annular fuel compact having the lowest maximum temperature, or tensile thermal stress, was determined for a given heat flux. The optimum volumetric fuel concentration in the compact was determined to be nearly independent of the over-all fuel fraction and geometry and to be approximately one-half when minimizing temperatures and one-third when minimizing stress in a $\text{UO}_2\text{-BeO}$ fuel body.

Since the over-all fuel volume fraction is usually small (at most 10%) the fueled region will be very thin and slab geometry approximations are justified. The following results are then easily derived. They may be shown to be also valid for a thin annular element cooled on both sides with an inner nonfueled region.

1. If a linear law of variation of thermal conductivity of the fuel compact (K_c) is assumed, then

$$K_c = xK_f + (1 - x)K_m,$$

where x is the volumetric fuel concentration in the compact and K_f and K_m are the conductivities of the fuel (and fertile) material and matrix material, respectively. The optimum concentration is found to be

$$x_0 = \frac{1}{2} \left[1 - \left(\frac{\bar{K}_f}{\bar{K}_m} \right) \right]^{-1},$$

where

$$\bar{K}_f / \bar{K}_m = \int_{T_s}^{T_{\max}} K_f(T) dT / \int_{T_s}^{T_{\max}} K_m(T) dT$$

is the ratio of conductivity integrals. T_s and $T_{\max}$ are the surface and maximum fuel compact temperatures, respectively. The ratio of heat flux to maximum heat flux for a given maximum temperature may be written

$$\frac{Q}{Q_0} = \frac{x}{x_0} \left(2 - \frac{x}{x_0} \right),$$

while the heat flux, when no matrix material is contained in the fuel compact, reads

$$Q_1/Q_0 = 4 \bar{K}_f/\bar{K}_m (1 - \bar{K}_f/\bar{K}_m) .$$

For UO_2 dispersed in BeO ,

$$\bar{K}_f/\bar{K}_m = 1/10 \text{ (approximately) ,}$$

$$x_0 = 0.555 ,$$

and

$$Q_1/Q_0 = 0.36 .$$

Usually the maximum volumetric fuel concentration in the fuel compact is limited by metallurgical or fabrication considerations. When $x = 0.25$, $Q/Q_0 = 0.7$, i. e., the heat flux is still approximately twice as large as for the case of pure fuel material. For a homogeneous fuel element

$$\frac{Q_\beta}{Q_0} = 4\beta \left(1 - \frac{\bar{K}_f}{\bar{K}_m} \right) \left[1 - \beta \left(1 - \frac{\bar{K}_f}{\bar{K}_m} \right) \right]$$

where β is the over-all volumetric fuel concentration. Even when $\beta = 1/10$, $Q_\beta/Q_0 = 0.328$ for $\bar{K}_f/\bar{K}_m = 1/10$, i. e., the heat flux in a homogeneous element is much smaller than could be obtained with a 25% volumetric fuel concentration in the compact.

2. Approximate formulas may be obtained by using average thermal conductivities when considering either average temperatures or thermal stresses, and for other laws of variations of thermal conductivity of the fuel compact. For instance, an approximate expression for the maximum thermal stress in a thin fuel body (of thickness e) cooled at the outside is

$$\frac{\sigma_{\max}}{\sigma_a} \cong \frac{Q}{6\pi\phi r} e ,$$

where $Q/2\pi r$ is the surface heat flux and $\phi = (1 - \nu) K \sigma / \alpha E$ is the thermal stress parameter. Using the correlation quoted in GA-5036, page 9, for BeO ,

$$\frac{\phi}{\phi_0} \cong 1 - \frac{3}{2} x ,$$

the ratio of heat flux to maximum heat flux for a fixed ratio of maximum stress to rupture modulus (σ_m/σ_a) becomes also

$$\frac{Q}{Q'_0} = \frac{x}{x'_0} \left(2 - \frac{x}{x'_0} \right),$$

where the optimum concentration is now $x'_0 = 1/3$. With $x = 0.25$, $Q = 15 Q'_0/16$ is very close to its maximum value, Q'_0 . Notice that the maximum heat per unit length is now

$$Q'_0 = \frac{2\pi\phi_0}{\beta} \frac{\sigma_{\max}}{\sigma_a},$$

where ϕ_0 is the stress parameter for pure BeO, while for given maximum temperature, it becomes

$$Q'_0 = 2\pi \frac{K_m (T_{\max} - T_s)}{\beta [1 - (K_f/K_m)]}.$$

If the subscript stands for matrix, the expression could be written

$$Q'_0 = \frac{2\pi}{\beta} \left(\frac{\sigma_{\max}}{\sigma_a} \right) \left[(1 - \nu) \frac{K\sigma}{\alpha E} \right]_m.$$

The ratio Q'_0/Q_0 is independent of the fuel loading (β):

$$\frac{Q'_0}{Q_0} = \frac{\sigma_{\max}/\sigma_a}{(T_{\max} - T_s)} \frac{\phi_0}{K_m} \left(1 - \frac{K_f}{K_m} \right).$$

For unfueled BeO at 1200°C , $\phi_0 \cong 10 \text{ w/cm}$ and $K_m \cong 0.2 \text{ w/cm-}^\circ\text{C}$, while $K_f/K_m \cong 0.1$ for UO_2 ; therefore, $Q'_0/Q_0 = 30/(T_m - T_s)^\circ\text{C}$ with $\sigma_{\max}/\sigma_a = 2/3$.

With $\beta = 1/10$, $Q'_0 \cong 420 \text{ w/cm}$, while $Q_0 = 2100 \text{ w/cm}$ for $T_m - T_s = 150^\circ\text{C}$, and $Q'_0/Q_0 = 1/5$. Thermal stresses usually determine the maximum permissible linear heat flux for fueled BeO bodies.

With 25% fuel concentration in the compact and a maximum flux of 400 w/cm, the maximum thermal stress is about two-thirds of the rupture modulus, and the maximum temperature drop is only about 40°C . Usually the maximum permissible fuel temperature will not be reached.

Conceptual Design of a Graphite-clad Fuel Element

It was shown in GA-4966 that the maximum heat flux obtainable with a fuel element cooled on both sides is much larger than for a cylindrical element cooled on the outside for the same maximum internal temperature and coolant conditions. In the study reported in GA-4966 the inner rod inside the annular element was not fueled. It is possible to obtain a still higher power density by using an inner rod consisting of an unfueled spine, an annular fuel compact, and a sleeve. Such an element is shown in Fig. 6, where the annular part of the element consists of two concentric sleeves between which are two fueled regions and an inner unfueled region. In that configuration, the maximum temperatures and thermal stresses are reduced in comparison with a homogeneous compact with fuel uniformly dispersed in a matrix (see the previous section, "Optimum Concentration in Fueled Region of Fuel Element Producing Maximum Heat Flux").

The dimensions of the fuel element were determined in the following manner: The total volume fraction occupied by the graphite sleeves was assumed to be about 25%, the thickness of the sleeves being 0.5 cm. The internal and external coolant voids were assumed to be 10% each, corresponding to a closely packed hexagonal lattice of fuel elements. The same pressure drops were assumed in the two channels for the same mass flows. About one-third of the total amount of BeO is fueled; this ratio derives from the need for 25 vol-% of fuel and fertile materials in the fueled BeO regions to minimize temperatures and thermal stresses and from the fact that the volume of fuel and fertile material must be approximately 8% of the total BeO volume from nuclear considerations. Thus, the dimensions shown on Fig. 6 are obtained where the over-all element diameter is 15 cm and the diameter of the inner rod is 6.65 cm.

With this geometry, for the same power density in the fueled regions, 46% of the heat is generated in the outer fuel annulus and removed in the outer coolant channel, 32% is generated in the inner annular fueled region, and 22% is generated in the inner rod. A more balanced design could be obtained by very slightly thickening the outer compact and slightly decreasing the innermost fuel compact, from 0.5 to 0.4 cm for instance.

A preliminary thermal study was performed for this fuel element, assuming temperature drops in the gaps between fuel compacts and sleeves to be equal to the temperature drops in the compact. Future studies will take into account the variation of gap sizes with temperature and lifetime in a fashion similar to that reported in GA-4451. The same mass flows were assumed in the two coolant channels, and therefore the coolant temperature rise is slightly higher in the inner channel.

Fig. 6--Conceptual design of an advanced beryllium oxide fuel element

The thermal conductivities were taken as 0.5 w/cm-°C for graphite, 0.33 for BeO, and 0.033 for heavy-metal oxides. The thermal-stress parameter $(1 - \nu)K\sigma/\alpha E$, was assumed to be 200 w/cm for graphite, 10 w/cm for BeO, and 6 w/cm for fueled BeO. Of the total heat, 10% was assumed to be generated in the unfueled BeO.

The Stanton number of the coolant $(h/\rho V C_p)$ was taken as 2×10^{-3} , while the coolant temperature rise was 400°C (in the inner channel) and the bulk coolant temperature was 700°C at the location of maximum fuel temperature; the fuel-element length was 400 cm. The temperature drops and maximum thermal stresses are given in Table 1 for a maximum power per unit length of fuel element of 8000 w/cm or a maximum power density of about 40 w/cm³. The hot-spot temperature is between 1500° and 1600°C. If the average power density is 20 w/cm³, approximately 800 such elements in a core 400 cm high and 450 cm in diameter could produce 1250 Mw(t) or approximately 500 Mw(e) at a net efficiency of 40%. The total temperature drop is higher for the rod than for the annular region (140°C compared with 89°C).

Table 1
TEMPERATURE AND STRESS DISTRIBUTIONS IN THE
GRAPHITE-CLAD COMPOSITE FUEL ELEMENT

	Temperature Drop, °C	Max Thermal Stress, σ_m/σ_a
Inner rod		
Spine	19	0.63
Compact	75	3.2
Sleeve	46	0.115
Annulus		
Sleeve	40	0.10
Compact	43.5	1.86
Spine	5.5	0.18
Compact	46.5	2.0
Sleeve	40	0.10

σ_a = ultimate tensile strength.

The thermal stresses are very small in the graphite sleeve (10% of ultimate strength), and are acceptable in the unfueled portions of the BeO. They are quite large in the fuel compacts, which means that cracking will occur at the hot spots. Unfueled BeO and graphite should be sufficient to

ensure structural integrity of the fuel element. By slightly reducing the thickness of the fueled region of the inner rod, at a maximum power density of 20 w/cm^3 there should be no overstressing of fueled BeO.

If 0.3-cm silicon carbide sleeves were used (silicon carbide is stronger than graphite, with a thermal stress parameter of 40 w/cm and a thermal conductivity of $0.75 \text{ w/cm-}^\circ\text{C}$) the stresses in the sleeve would be about 30% of the ultimate strength at a maximum power density of 40 w/cm^3 . The volume fraction of silicon carbide would be only about 15%. A 0.3 cm-silicon carbide sleeve absorbs approximately the same amount of neutrons as 3 mils of Hastelloy-X.

MATERIALS

Development of UO_2 - ThO_2 -BeO Fuel Bodies

In the investigation of the development of UO_2 - ThO_2 -BeO fuel bodies, GC-grade BeO was included in the investigation in an effort to reduce the cost of ceramic fuel. Raw materials now on hand for use in the investigation consist of:

1. Approximately 14 kg of normal ceramic-grade UO_2 powder.
2. Approximately 50 kg of UOX- and GC-grade BeO powder.
3. Approximately 30 kg of ThO_2 plus an additional 34 kg being held in reserve for emergency use.

The work to develop a superior ceramic-type fuel continued during the quarter with extensive effort being applied to redetermine the sintered properties of GC- and UOX-grade BeO with and without flux additions. Variables being studied, as part of this investigation involving the basic components of the ceramic-type fuels, are listed below:

1. BeO studies are being conducted to:
Re-establish the sintered bulk densities of as-received and ball-milled UOX- and GC-grade beryllia.

Determine the effects on the sintered bulk density of GC- and UOX-grade BeO of the addition of MgO (0.25 to 0.5 wt-% and ThO_2 (0.25 to 64 wt-%).

Determine the effect on the sintered bulk density of BeO pellets fabricated from powder mixtures of GC- and UOX-grade beryllia as part of the effort to reduce the over-all fuel cost.

2. Thorium and uranium oxide studies include efforts to:
Establish the compaction and sintering properties of 10/1 and 20/1 mixtures of ThO_2 and UO_2 .

Establish the compaction and sintering properties of ceramic fuel bodies fabricated from each of the following combinations of ThO_2 , UO_2 , and BeO : (1) coarse UO_2 particles dispersed in a continuous matrix of BeO and ThO_2 ; (2) coarse particles fabricated from a mixture of UO_2 and ThO_2 dispersed in a continuous matrix of BeO ; (3) coarse particles fabricated from a mixture of UO_2 and ThO_2 dispersed in a continuous matrix of BeO fluxed with small additions of ThO_2 or MgO as sintering aids; (4) coarse UO_2 particles and ThO_2 particles dispersed in a continuous matrix of BeO ; and (5) a homogeneous mixture of fine BeO , UO_2 , and ThO_2 . In each of these combinations of BeO , UO_2 , and ThO_2 , milled and unmilled, fluxed and unfluxed, GC- and UOX-grade materials are incorporated in the fuel pellet to determine their effects on the sintered bulk density and on the ease of fabrication.

3. Niobium-coated- UO_2 -particle studies will:
Develop fugitive-type coating for the niobium-coated fuel particles to compensate for shrinkage of the matrix material during the sintering process.

Fabricate ceramic fuel bodies consisting of niobium-coated UO_2 particles dispersed in a continuous matrix of BeO and ThO_2 . In addition to this program, niobium-coated UO_2 particles were received, and means are being developed to incorporate them in a dense beryllia material.

4. Sol-gel studies are being conducted to:
Establish the compaction and sintering properties of as-dried and fired sol-gel.

Establish the density and shrinkage of sol-gel particles as a function of sintering temperature, and incorporate prefired sol-gel particles of compatible shrinkage into GC-grade and UOX-grade BeO pellets.

Establish the sintering properties of homogeneous fuel pellets fabricated by one of the following methods:

- As-received UOX- BeO plus dry sol-gel fines.
- As-received GC- BeO plus dry sol-gel fines.
- Ball-milled mixture of UOX- BeO and dried sol-gel.

Ball-milled mixture of GC-BeO and dried sol-gel.
 As-received UOX-BeO plus fired sol-gel fines.
 As-received GC-BeO plus fired sol-gel fines.
 Ball-milled mixture of UOX-BeO and fired sol-gel.
 Ball-milled mixture of GC-BeO and fired sol-gel.

Establish the sintering properties of seeded fuel pellets fabricated by one of the following methods:

As-received UOX-BeO plus dry sol-gel seeds.
 As-received GC-BeO plus dry sol-gel seeds.
 Ball-milled UOX-BeO plus dry sol-gel seeds.
 Ball-milled GC-BeO plus dry sol-gel seeds.
 As-received GC-BeO plus fired sol-gel seeds.
 Ball-milled GC-BeO plus fired sol-gel seeds.

Table 2 shows the results obtained to date on the sintered densities of pellets fabricated from ball-milled beryllia and as-received beryllia. UOX-grade BeO shows an increase of 11% in sintered bulk density whereas the GC grade shows an increase of 28% as a result of ball-milling in distilled water for 1 hr.

Table 2
COMPARISON OF PELLETS FABRICATED FROM
GC-GRADE AND UOX-GRADE BERYLLIA

BeO Material ^a	Flux Addition		Shrinkage, %	Sintered Density	
	Material	wt-%		g/cm ³	% theo
As-received UOX grade	----	----	13.3	2.64	87.7
As-received GC grade	----	----	6.4	2.10	70.0
As-received GC grade	MgO	0.25	12.1	2.51	83.4
As-received GC grade	MgO	0.5	14.0	2.70	89.7
Ball-milled UOX grade	----	----	16.6	2.97	98.7
Ball-milled GC grade	----	----	15.7	2.95	98.0

^a All pellets were compacted at 21,500 psi and sintered in hydrogen for 2 hr at 1,650°C.

Table 3 shows the effect of mixing various percentages of UOX- and GC-grade BeO powders together.

Table 4 shows the effect of mixing from 0.25 to 64 wt-% ThO₂ in GC-grade BeO. All components were ball-milled together for 1 hr prior to being fabricated into pellets. Sintered densities ranged from 94.1% 98% of theoretical.

Table 3

EFFECTS ON SINTERED DENSITY OF MIXING BALL-MILLED
UOX- AND GC-GRADE BERYLLIA

Material ^a		Shrinkage, %	Sintered Density	
GC-BeO, wt-%	UOX-BeO, wt-%		g/cm ³	% theo
15	85	14.5	2.89	96.6
30	70	14.0	2.93	97.3
60	40	14.7	2.95	98.0
75	25	15.2	2.94	97.7

^a All pellets were compacted at 21,500 psi and sintered in hydrogen for 2 hr at 1,650°C.

Table 5 shows the effect of incorporating fully sintered sol-gel fines into BeO fuel pellets. The resulting high density may be somewhat misleading because of the initial high density of the sol-gel material, which was already nearly of theoretical density.

Table 6 shows the effect of incorporating as-received sol-gel fines into BeO fuel pellets.

Table 4

EFFECTS OF THORIUM OXIDE ADDITIONS ON SINTERED
DENSITY OF BALL-MILLED GC-GRADE BERYLLIA

GC-Grade BeO, wt-%	ThO ₂ , wt-%	Shrinkage, %	Sintered Bulk Density	
			g/cm ³	% theo
99.75	0.25	15.2	2.92	96.8
99.50	0.50	15.2	2.95	97.7
99	1.0	15.2	2.97	98.0
98	2.0	15.2	2.98	97.7
96	4.0	15.0	2.98	96.1
92	8.0	15.5	3.10	97.2
84	16.0	15.7	3.28	96.8
68	32.0	15.5	3.73	96.3
50	50.0	16.2	4.39	94.8
36	64.0	15.5	5.13	94.1

Table 5

SINTERED-DENSITY RESULTS OF PELLETS FABRICATED FROM
20-WT % FULLY SINTERED SOL-GEL FINES AND 80 WT-% BeO

Test No.	Grade of BeO	Green Density, % theo	Shrinkage, %	Sintered Bulk Density	
				g/cm ³	% theo
39	GC	58.0	15.2	3.32	96.0 ^a
40	UOX	59.1	14.5	3.30	96.4 ^a
43	UOX	60	14.1	3.38	96.6 ^b
44	GC	61.4	10	2.98	85.1 ^b

^a All components were ball-milled together for 1 hr.

^b All components were manually mixed together.

Table 6

SINTERED-DENSITY RESULTS OF PELLETS FABRICATED
FROM AS-RECEIVED SOL-GEL FINES AND BERYLLIA

Test No.	Grade of BeO	Green Density, % theo	Shrinkage, %	Sintered Bulk Density	
				g/cm ³	% theo
47	UOX	57.7	15.7	3.29	94.0 ^a
48	GC	58.6	15.7	3.28	93.7 ^a
45	UOX	58.3	15.2	3.33	95.1 ^b
46	GC	59.7	9.0	2.73	78.0 ^b

^a All components were ball-milled together for 1 hr.

^b All components were manually mixed together.

Table 7 shows the effects upon the "bulk" density of fabricating pellets from already fully sintered sol-gel fines. The bulk density is misleading in this case because the individual fully sintered sol-gel particles are of nearly theoretical density prior to being compacted into a pellet, and little tendency exists for them to congeal together during the sintering operation.

Lithium-6 in Pellets from BeO Capsule MGCR-2

During the quarter the analytical procedure for determination of Li⁶ in beryllia was improved, and the analyses reported in the previous quarterly report were redetermined. Final Li⁶ concentrations as determined

by the improved procedure are given in Table 8, together with calculated values for comparison. As can be seen, the experimentally determined and the calculated numbers agree well.

Table 7

SINTERED-DENSITY RESULTS OF SOL-GEL PELLETS
FABRICATED FROM FULLY SINTERED SOL-GEL FINES

Test No.	Material	wt-%	Green Density, % theo	Shrinkage, %	Sintered Bulk Density	
					g/cm ³	% theo
41	---	-	78.3	1.2	8.31	82.5
42	LiF	1	79.8	0.6	8.28	82.2

Table 8

LITHIUM-6 CONCENTRATION IN MGCR-2 BeO

Sample No.	Location in MGCR-2	Measured $\mu\text{g Li}^6/\text{g BeO}$	$\mu\text{g Li}^6/\text{cm}^3 \text{BeO}^a$	Calculated $\mu\text{g Li}^6/\text{cm}^3 \text{BeO}$
50030	Center pellet	7.3 $\pm$ 0.3	18.2	20.4
50031	Inner ring	7.0 $\pm$ 0.2	17.6	20.5
50032	Outer ring	7.7 $\pm$ 0.4	20.8	20.75
50092	Center pellet	10.1 $\pm$ 0.2	26.6	24.75
50093	Inner ring	9.9 $\pm$ 0.2	25.2	24.85
50094	Outer ring	10.6 $\pm$ 0.3	26.0	25.1

^aConversion of measured values.

Diffusion of Lithium-6 in BeO

The investigation of the diffusion of Li^6 in BeO continued during the quarter. Results from the method reported in the previous quarterly report have not as yet led to a firm estimation of the diffusion coefficient constant in BeO.

Experiments with a different analytical procedure are underway. In this procedure, BeO cylinders are annealed in a helium atmosphere and the released Li^6 is collected on a cold finger and then deposited on an alumina surface for irradiation. Results from such experiments are given in Table 9 and are plotted in Fig. 7. If the release value is corrected for the collection efficiency of the cold finger, an estimated diffusion coefficient, $D' = D/a^2$, of $1.3 \times 10^{-7} \text{ sec}^{-1}$ is obtained.

Fig. 7--Percentage of released Li^6 (from MGCR-2 capsule) collected on cold finger versus the square root of time

Table 9

LITHIUM-6 RELEASE FROM IRRADIATED BeO

BeO cylinder	80% density (approx)
Weight before annealing	559.2 mg
Weight after annealing	555.1 mg
Li ⁶ content before annealing	7.3 μ g Li-6/g BeO
Helium purge	55 cm ³ /min
Temperature	1390 $\pm$ 5 ^o C

Annealing Time, min	Li ⁶ at Cold Finger, μ g	Total Release of Li ⁶	
		μ g	%
1346	0.128	0.128	3.14
2690	0.021	0.149	3.65
4037	0.007	0.156	3.83

Four-cell Test Capsule 2F1

Capsule GA3-040-2F1 was shipped to the MTR during the previous quarter and inserted in irradiation position A-20. After approximately two weeks of operation, a large leak developed in the lead tube, necessitating repair in-place and subsequent reactor shutdown for nondestructive transfer of the capsule to the reactor canal. A new lead tube was installed and the capsule, reinserted in the reactor, operated normally with a temperature in cells 1 and 2 of 2700^oF and in cell 3 of 2800^oF. Cell 4 was below design temperature. On June 7, 1964, the lead tube began losing pressure and bubbles started flowing from the capsule. As a result of subsequent examination, the capsule has been permanently discharged from the reactor.

One-cell Test Capsule F5

Irradiation capsule GA4-F5 will test the effects of irradiation on UO₂-ThO₂-BeO fuel pellets. Capsule F5 was originally planned as a static capsule to back up sweep capsule GA3-040-2F1. However, capsule F5 will be designed as a one-cell sweep capsule and will utilize the equipment (gas-sampling station) from 2F1 at the MTR. Design and construction of F5 was initiated during the quarter.

3. PLUTONIUM UTILIZATION INVESTIGATION

PHYSICS

Replacement Core for EBOR

Preliminary investigations of the feasibility of a 10-Mw(e) plutonium-fueled core as a second core for EBOR have been completed. Such a core would have a lifetime equal to or somewhat greater than that of the present core, would emulate many of the features of a large plutonium-fueled power reactor, and would demonstrate the Phoenix-fuel effect. In GA-5036 it was pointed out that such a concept appears to be feasible, provided the core can be constructed with a minimum amount of metal. Cladding of the fuel elements with metal, as in the present EBOR core, would not lead to a successful test.

A more detailed examination of the characteristics of a 3-ft core with 18.3% Pu²⁴⁰ (Fig. 14 of GA-5036) shows some of the reasons for the behavior of a typical Phoenix-fueled core. The time-dependent neutron balance for this case is shown in Fig. 8. The production of source neutrons per fuel absorption (η_{eff}) increases with lifetime because of the increasing number of absorptions in Pu²⁴¹ relative to Pu²³⁹ and because Pu²⁴¹ has a higher value of η than Pu²³⁹. Simultaneously, the absorptions in Pu²⁴⁰ decrease with time. Since the absorptions in fission products, Pu²⁴², and Li⁶ increase with time, a balance can be achieved in which the net increase in nonfuel absorptions is equal to the increase in available source neutrons, resulting in a constant excess of source neutrons, which leads to the constant reactivity-lifetime characteristics associated with Phoenix-fueled cores. End of life comes when the fuel depletion is so great that the reactor expends nearly all of its fuel. Figure 9 shows the plutonium composition as a function of time for this same case. The fuel content of the core is reduced by burnup to less than one-third of the initial value. At end of life, the plutonium is nearly 30% nonfissile, compared with 20% nonfissile at the beginning of life. It is therefore unlikely that the plutonium can be recycled, and in cases where burnup is even higher, the reprocessing of fuel may be eliminated entirely because of a lack of economic justification. The decrease in Pu²⁴⁰ absorptions shown in Fig. 8 is partly the result of the depletion of Pu²⁴⁰ and partly the result of softening of the neutron spectrum caused by fuel depletion. The softening spectrum is illustrated by the change in the median fission energy with lifetime as shown in Fig. 10. It is evident that the median fission energy is in the

Fig. 8--Neutron balance for 3-ft core containing 18.3% Pu^{240}

Fig. 9--Plutonium composition versus life-time for 3-ft core containing 18.3% Pu^{240}

Fig. 10--Variation in median fission energy with lifetime for 3-ft cores

Fig. 11--Effect on reactivity-lifetime curves for 3-ft cores of adding niobium coating, thorium oxide, and changing specific power in cores containing 18.3% Pu^{240}

vicinity of the low-lying Pu^{239} resonance where η is at a minimum (illustrated by Fig. 3.7 of GA-4165). A decrease in the median fission energy or a softening of the spectrum results in an increase in η , which augments the increase in η from the buildup of Pu^{241} , as shown in Fig. 8.

The results presented in the previous quarterly progress report (GA-5036) (Figs. 14 to 25) were derived for reactors of an initial specific power of 500 kw/kg of fissile material, which matches the desired values for larger power reactors. At this specific power, the core lifetime is 4 to 5 yr, which is about 3 times greater than the design lifetime of EBOR. From the time-dependent neutron balance shown in Fig. 8, it is seen that to reduce the lifetime but still maintain the flat reactivity characteristic, it is necessary to increase the specific power so that full burnup is reached sooner. As a result of increasing specific power, the neutron spectrum becomes softer, the value of η increases, the absorptions in Pu^{240} decrease, and the reactivity therefore increases. The spectrum-induced changes in η and Pu^{240} absorptions become less, and for large changes in specific power an increase in the Pu^{240} concentration may be necessary to achieve the flat reactivity-lifetime characteristic. In order to reduce excess reactivity, the excess neutrons must be absorbed in some nondepleting material so that the flatness of the curve is not altered. Thorium is added to the fuel to perform this function. In the EBOR replacement core, only a nominal amount of U^{233} is formed, but the U^{233} production available in a larger reactor would be appreciable. The reactivity-lifetime effects that derive from changing the specific power and adding thorium are shown in Fig. 11. Note that at an initial specific power of 1000 kw/kg and a BeO/Th ratio of 625, the lifetime is just over 600 days, or approximately 2 yr, with a maximum excess reactivity of 1.2%. It should be mentioned that all of the curves in this section were derived using GGC-II data as discussed in the previous quarterly progress report.

In the calculations above, the core configuration was assumed to be composed of closely spaced cylindrical elements on a triangular pitch, conforming closely to larger core designs. Such an arrangement of fuel elements in the EBOR pressure vessel would require the design and fabrication of a bottom support grid plate and plenum that would fit over or replace the present grid plate and direct the coolant into the proper channels. In addition, control rods and control-rod drives would require extensive modification. The alternative is to design the fuel elements to fit the existing support-plate configuration. The void volume, in a configuration in which the fuel elements touch and are arranged on a square pitch is 27.2% of the total (including the control-rod voids), compared with 9.1% for a triangular pitch without allowance for control rods. The average power density can be reduced, however, and the heat generation rate in the fuel element can still be matched to the prototype. A 3.5-ft core operating at a power density of 11.2 kw/liter and producing 10.7 Mw(t) of power

has a fuel heat-generation rate equivalent to that of the triangular-pitched core at a power density of 14 kw/liter. The effect of increasing the void volume is to harden the neutron spectrum, so that less Pu^{240} is required to maintain the flat reactivity-lifetime characteristic. Figure 12 shows the results of calculations parallel to those of Fig. 11 but with the increased void volume, lower over-all power density, and a Pu^{240} content of 15.7%. Again, at 1000 kw/kg a lifetime of nearly 700 days is attainable with a maximum excess reactivity of 1.3% using a BeO/Th ratio of 350. These parameters will serve as a starting point for the design of the replacement core.

MATERIALS

Fuel Fabrication Development

The fabrication of PuO_2 -BeO and PuO_2 - ThO_2 -BeO fuel bodies was discussed with General Electric Company Hanford Plant personnel. The application of the Nupak (Dynapak) machine to the production of dense dispersion-type oxide fuel bodies appears to be possible and may have advantages over the more conventional fabrication processes.

Silicon Carbide as Sleeve Material

A review of recent properties data has suggested silicon carbide as a possible fuel-element cladding material. The previous quarterly report included a preliminary appraisal of silicon carbide as a sleeve material. It was pointed out that the thermal stress properties of SiC exceed those of BeO. Furthermore, SiC is compatible with air, carbon dioxide, steam, and hydrogen at elevated temperatures; it is impervious and should have good radiation stability because of its isotropic structure. Its microscopic thermal-neutron cross section is larger than that of BeO or graphite but much lower than that of stainless steel or Hastelloy-X.

Two types of silicon carbide were examined: (1) KT silicon carbide, made by the Carborundum Company; and (2) vapor-deposited silicon carbide, made by the San Fernando Laboratories. The KT silicon carbide is fabricated by an extrusion process, and has been made in diameters up to 4 in. with 1/4-in. -wall thickness and in lengths up to 4 ft. It is a dense, impervious material. However, the dimensional tolerances are poor, since there is approximately a 1/4-in. bow on a 4-ft-long tube, and the tube is approximately 60 mils out-of-round in cross section. The vapor-deposited silicon carbide has excellent dimensional tolerances, as well as high density and low permeability.

Fig. 12--Reactivity-lifetime characteristics of 3.5-ft cores with EBOR voids at 10.7 Mw(t) and containing 15.7% Pu²⁴⁰

4. EBOR TEST MODULE

During the quarter, a preliminary study was initiated for design and development of an advanced beryllia module for testing in EBOR. The essential steps of the work are described in the following paragraphs.

1. Parametric studies completed to date on fuel elements utilizing the $\text{BeO-ThO}_2\text{-UO}_2$ and the $\text{BeO-ThO}_2\text{-PuO}_2$ fuel cycles have been sufficiently favorable to justify a program of theoretical and experimental investigation for development of an EBOR test module based on an initial $\text{BeO-ThO}_2\text{-UO}_2$ fuel cycle.

An outline of the principal areas of interest related to this investigation is shown on Fig. 13. As used on the figure, ABE and ABE(T) are respectively abbreviations for advanced beryllia element and advanced beryllia element test.

In future work the scope of investigation in each area will be limited to the development of data specifically required for the test-element program.

2. An investigation was made to determine the effect of a gap between the fuel body and sleeve on the thermal performance of the test module. The calculational model chosen consisted of an annular $\text{BeO-UO}_2\text{-ThO}_2$ fuel body with a BeO spine and a graphite sleeve, which was essentially the center section of the fuel element shown in Fig. 6. The outside diameter was 7.45 cm comprising a 4.65-cm spine, a 0.5-cm-thick annular fuel compact, a 0.5-cm-thick graphite sleeve and a 0.4-cm-wide coolant passage. EBOR operating coolant conditions (coolant inlet temperature of 770°F) and an axial power profile corresponding to the condition after 2500 hr of power operation were assumed in the calculation. Figure 14 shows the maximum permissible power density versus a helium hot gap for various assumed maximum fuel-body temperatures. It can be seen from the figures that the allowable power density is sensitive to the gap width. The significance of this fact is that the ultimate dimensional changes in the fuel body resulting from irradiation determine the size of the original fuel body.

These fuel-body dimensional changes result from a large number of other variables, including manufacturing processes, component size, temperature, and flux. In a fuel-element design consisting of a BeO

Fig. 13--Flow diagram showing principal areas of interest of the advanced beryllia test-module development program

Fig. 14--Power density versus gap width for various fuel temperatures

spine-fuel compact-graphite sleeve arrangement the graphite is comparatively stable, but the fuel compact grows appreciably over its lifetime. It is assumed that the graphite sleeve cannot withstand a tensile mechanical load, therefore a finite gap must be provided at the beginning of life to provide room for the fuel compact to expand during operation. Reference 2 indicates diametral growth for beryllia ranging from approximately 0.25% to 3%. Assuming diametral changes in this range for the fuel body, the allowable fuel-body diameter is given in Table 10 for two power densities and two gap conductivities.

Table 10
EFFECT OF DIMENSIONAL GROWTH ON ALLOWABLE
FUEL-BODY DIMENSIONS

Average Power Density, kw/ℓ	Gap Conductivity		Hot-gap Change, ^a mils	Diametral Growth, %	Fuel-body Diameter	
	Btu/ hr-ft-°F	w/ cm-°C			in.	cm
15	0.28	0.00485	15	0.25	6.0	15.2
				0.5	3.0	7.62
				1	1.5	3.81
				2	0.75	1.91
				4	0.325	0.95

^a Beginning-of-life to end-of-life change where $T_{\max}$ is 1600°C (2912°F).

3. A preliminary investigation was made to determine the consequence of graphite burning on adjacent fuel elements. In some postulated EBOR accidents the core is cooled by means of the emergency air cooling system. The graphite reacts exothermically with the oxygen in the air at a rate dependent on the temperature. Using the reaction-rate data shown in Fig. 15, (3-5) calculations showed that the oxygen in the air is consumed after the coolant has proceeded 24 in. along the channel and that the burning time is approximately 30 min.

A heat-transfer analysis was performed using the code RAT, a two-dimensional generalized heat-transfer program. The calculational model was arranged in cylindrical geometry with the axis of symmetry at the centerline of the graphite sleeve. The adjacent elements were lumped into annular rings representing the BeO block, the shroud tube, and fuel. The results of the analysis are shown in Fig. 16, which shows the shroud-tube temperature versus time during the graphite burning period. It can be

Fig. 15--Graphite reaction rate

Fig. 16--Temperature distribution during burning period

seen that the maximum shroud-tube temperature is approximately 1650°F (the maximum shroud-tube temperature is approximately 1400°F in the original EBOR configuration). In addition, the results show that the heat flow from the burning graphite to the fuel element causes a temperature gradient in the BeO square annular block. Because of the cylindrical geometry used for this analysis, the quantitative results are only approximate; however, the maximum radial temperature differential noted (35°F) is comparable in magnitude to the radial temperature differential in the BeO blocks adjacent to the control blade passage under EBOR design conditions.

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